Coherent single cycle pulses with MV/cm field strengths from a relativistic transition radiation light source

Matthias C. Hoffmann¹, Sebastian Schulz², Stephan Wesch², Steffen Wunderlich², Andrea Cavalleri^{1,3} and Bernhard Schmidt²

¹ *Max Planck Research Department for Structural Dynamics, University of Hamburg - CFEL, Hamburg, Germany*

² *Deutsches Elektronen-Synchrotron (DESY), Hamburg, Germany*

³ *Department of Physics, Clarendon Laboratory, University of Oxford, United Kingdom*

Abstract— THz pulses with energies up to 100 μJ and corresponding electric fields up to 1 MV/cm were generated by coherent transition radiation from 500 MeV electron bunches at the free electron laser FLASH. The pulses were characterized in the time domain by electro-optical sampling by a synchronized femtosecond laser with jitter of less than 100 fs. High THz field strengths and quality of synchronization with an optical laser will enable observation of nonlinear THz phenomena.

Intense single-cycle THz pulses are increasingly being used to observe extreme nonlinear optical phenomena at high field strengths [1-3]. These pulses can be obtained on the table-top by optical rectification of femtosecond laser pulses and can reach pulse energies on the level of 10's of μJ [4]. Even higher pulse energies and fields can potentially be reached by accelerator-based sources [5, 6]. Coherent transition radiation (CTR) is emitted when an electron bunch crosses the boundary between two media [7, 8]. The energy of the pulses scales with the average line charge density ("peak current") of the bunch squared and their frequency content covers the THz up to infrared region of the electromagnetic spectrum depending on the electron bunch duration. Since the driving linear accelerators for high gain free electron lasers (FEL) are built to boost the peak current to several kA, they are well suited to drive powerful CTR sources. A number of high-gain X-ray FEL facilities (LCLS, ELETTRA, PAL) are now developing a complementary THz research program with short broadband THz pulses. At the soft x-ray free-electron

laser FLASH [9] transition radiation is generated from collision of electrons bunches with energies up to 1.2 GeV with a metal screen and then guided through an evacuated beam line [10] with 20 m length into a user accessible lab. While the main purpose of this beam line is high-resolution diagnostics of the longitudinal electron bunch characteristics [11], it can also serve as a powerful and broadband source for experiments with sub-picosecond single cycle pulses. The pulses obtained have energies exceeding 100 μJ and span a frequency band of 200 GHz to 100 THz. Here we determine the temporal structure and the electric field of the THz pulse below 10 THz directly in the time domain using electro-optic sampling with femtosecond laser synchronized to the electron bunches. The availability of optical probe lasers synchronized to this intense THz source enables THz pump optical probe measurements.

The CTR source at FLASH is located in a straight section between the superconducting linear accelerator and the undulator. The last electron bunch within a sequence of up to 800 bunches with a spacing of 1 μs can be deflected onto an off-axis CTR target by a fast kicker magnet without interfering with routine FEL user operation at the x-ray undulator downstream. The CTR radiation is produced on a metal screen of 150 nm aluminum on a 380 μm silicon wafer within the high vacuum part of the FLASH beam line and then transported to a dedicated optical laboratory outside the accelerator tunnel. In order to obtain the largest possible bandwidth, a diamond window with a 20 mm diameter and 0.5 mm thickness is used to separate the high vacuum inside the accelerator tube from the beam line. The beam line itself is evacuated to better than 0.1 mbar to prevent atmospheric absorption. The transport tube has a minimum diameter of 200 mm to maintain low frequencies. A detailed description of the beam transport theory of CTR generation at FLASH can be found in Ref. [10].

In order to enable direct characterization of the THz field in the time domain it is vital to use a synchronized laser system with a jitter on the order of 100 fs or less relative to the electron bunch. For our experiments a Ti:sapphire femtosecond oscillator is synchronized to the FLASH master oscillator of the accelerator by high frequency to baseband mixing (Fig. 1). An electrical frequency comb is generated

from the laser pulse train by a fast photodiode with >10 GHz bandwidth. The 16th harmonic of the laser repetition rate is filtered out, amplified and used as the local oscillator input of a frequency mixer. The RF signal provided by the master oscillator passes a digitally-controllable vector modulator which enables to shift the laser phase with respect to the electron beam. The down-mixed signal is processed in a DSP controller, acting on a piezo stack attached to a laser cavity folding mirror, thus providing synchronization to the accelerator. Since a high harmonic of the repetition rate is used for precise synchronization, the phase-locked loop (PLL) can lock on at 16 possible zero-crossings which results in an ambiguity of the relative timing between the laser and THz pulses. To overcome this, a similar phase detector operating at the fundamental repetition rate of the laser is installed. Its output is also fed into the PLL and the software automatically locks the laser first at the fundamental frequency to ensure the correct timing and then switches over to the higher frequency for a tighter synchronization. Using this scheme, a synchronization accuracy of better than 40 fs (RMS) can be achieved between the laser oscillator and the master oscillator.

Figure 1: Laser synchronization scheme used at the CTR beam line laser lab at FLASH. The 16th harmonic of the laser repetition rate and a phase shifted signal from the 1.3 GHz master oscillator controlling the electron gun are used as input of a frequency mixer. The down-mixed signal is used to acting back on a cavity folding mirror via a piezo stack. For details see text.

It should be noted that the actual timing uncertainty between electron bunch and laser oscillator is larger due to additional jitter introduced by the acceleration process and estimated to be on the order of 100 fs.

THz pulses with up to 100 μJ energy were produced by electron bunches with 0.6 nC charge and a few hundred fs duration at a repetition rate of 10 Hz. Since the CTR beamline is running parasitically to normal user operation for FEL experiments, no specific tuning of bunch charge or duration for optimum THz output was carried out. The THz pulse energy was measured by a calibrated pyroelectric detector. THz radiation was focused by a 3 inch diameter 3 inch focal length off axis parabolic mirror inside a vacuum vessel where the beam profile was characterized using a pyroelectric camera, yielding a minimum spot size of 1 mm FWHM at the focus. The THz electric field was measured by electro-optical sampling [12] using 0.2 mm $\langle 110 \rangle$ cut GaP crystal mounted on inactive $\langle 100 \rangle$ substrate. The polarization state of the femtosecond laser pulses transmitted through the GaP crystal was analyzed by a combination of quarter wave plate and Wollaston prism. The laser light was detected by a pair of balanced 1 ns risetime photodiodes and the individual fs pulses were gated electronically to select for temporal coincidence with the THz pulses. Temporal delay in steps of 33 fs was achieved by changing phase of the vector modulator. For the highest field strengths, the THz radiation had to be attenuated with a pair of wire grid polarizers in order to prevent over-rotation of the probe ellipticity inside the detection crystal.

A typical field trace at 80 μJ pulse energy and the corresponding frequency spectrum obtained by Fourier transformation is shown in Figure 2. For this measurement 30 pulses were averaged at every time step. The field strength was calculated from the known electro-optical coefficient of GaP taking into account Fresnel losses but neglecting phase mismatch of laser and THz pulse inside the GaP crystal. The intensity modulation in a balanced detection scheme caused by a THz field E_{THz} is

$$\sin^{-1}(\Delta I / I) = 2\pi n_0^3 r_{41} t_{\text{GaP}} E_{\text{THz}} L / \lambda_0 \quad (1)$$

where $n_0=3.193$ is the refractive index of GaP at 800 nm, $L=0.2$ mm the thickness of the crystal, $\lambda_0=800$ nm the probe wavelength, r_{41} the electro optic coefficient and t_{GaP} is the Fresnel coefficient for reflective loss at the GaP crystal. The value for the electro optic coefficient is $r_{41}=0.88$ pm/V [13]. In our experiments we observed values up to $\Delta I/I=(I_1-I_2)/(I_1+I_2)=0.66$ for signals I_1 and I_2 detected by the photo diodes located after the Wollaston prism at peak THz field. The THz field was attenuated by factor of 0.5 using a pair of linear polarizers to avoid over-rotation of the phase picked up by the probe pulse in the electro-optic crystal.

Figure 2: Single cycle THz field with MV/cm field strength and corresponding amplitude spectrum (inset) as characterized by electro-optical sampling in a 0.2 mm GaP crystal.

Figure 2 shows the measured THz field strength vs. time delay. The pulse is clearly single cycle with peak field strength of 700 kV/cm while the spectrum obtained by Fourier transformation shows frequency components extend up to 4 THz. The bandwidth of our measurement is limited by the timing jitter between the electron bunches and the femtosecond laser which is on the order of 100 fs. When taking into account corrections for phase mismatch in the detection crystal, transmission loss in the two polarizers and signal reduction due to averaging because of synchronization jitter, the peak field strength is estimated to exceed 1 MV/cm. This is consistent with the value obtained from spot size and total energy

measurements. By inserting bandpass filters (QMC Instruments), narrowband THz pulses were generated. Figure 3 shows the electro-optical signal for a pulse with 2 THz center frequency and 0.5 THz spectral bandwidth. The field strength still exceeds 100 kV/cm.

Figure 3: Narrowband THz pulses can be obtained by using a bandpass filter with center frequency at 2 THz.

In summary, the CTR beam line at FLASH delivers single cycle THz pulses with field strength on the order of 1 MV/cm in a table-top user laboratory. These pulses have larger bandwidth and almost one order of magnitude more energy than similar pulses obtained by optical rectification [4]. The THz field can be directly characterized in the time domain using a femtosecond laser synchronized with 100 fs jitter. Further increases in THz field strength are expected from optimizing compression and energy of the electron bunches. Improvements in timing accuracy to below 10 fs [14] are possible by synchronizing to an optical reference [15] with a cross-correlation technique and by post-processing using data from bunch arrival monitors [16]. This unique combination of extreme intensity THz fields from an accelerator with ultrafast timing will allow for observation of novel nonlinear THz phenomena.

REFERENCES

1. T. Kampfrath, A. Sell, G. Klatt, A. Pashkin, S. Mahrlein, T. Dekorsy, M. Wolf, M. Fiebig, A. Leitenstorfer, and R. Huber, "Coherent terahertz control of antiferromagnetic spin waves," *Nat Photon* 5, 31-34 (2010).
2. M. C. Hoffmann, N. C. Brandt, H. Y. Hwang, K.-L. Yeh, and K. A. Nelson, "Terahertz Kerr effect," *Applied Physics Letters* 95, 231105-231103 (2009).
3. A. Dienst, M. C. Hoffmann, D. Fausti, J. C. Petersen, S. Pyon, T. Takayama, H. Takagi, and A. Cavalleri, "Bi-directional ultrafast electric-field gating of interlayer charge transport in a cuprate superconductor," *Nat Photon* 5, 485-488 (2011).
4. M. C. Hoffmann, and J. A. Fülöp, "Intense ultrashort terahertz pulses: Generation and applications," *Journal of Physics D* 44, 083001 (2011).
5. G. L. Carr, M. C. Martin, W. R. McKinney, K. Jordan, G. Neil, R., and G. P. Williams, "High-power terahertz radiation from relativistic electrons," *Nature* 420, 153-156 (2002).
6. G. P. Williams, "Filling the THz gap - high power sources and applications," *Rep. Prog. Phys.* 69, 301-326 (2006).
7. U. Happek, A. J. Sievers, and E. B. Blum, "Observation of coherent transition radiation," *Physical Review Letters* 67, 2962 (1991).
8. Y. Shibata, K. Ishi, T. Takahashi, T. Kanai, M. Ikezawa, K. Takami, T. Matsuyama, K. Kobayashi, and Y. Fujita, "Observation of coherent transition radiation at millimeter and submillimeter wavelengths," *Physical Review A* 45, R8340 (1992).
9. J. R. Schneider, "FLASH - from accelerator test facility to the first single-pass soft x-ray free-electron laser," *Journal of Physics B: Atomic, Molecular and Optical Physics* 43, 194001 (2010).
10. S. Casalbuoni, B. Schmidt, P. Schmüser, V. Arsov, and S. Wesch, "Ultrabroadband terahertz source and beamline based on coherent transition radiation," *Physical Review Special Topics - Accelerators and Beams* 12, 030705 (2009).
11. S. Wesch, B. Schmidt, C. Behrens, H. Delsim-Hashemi, and P. Schmüser, "A Multi-Channel THz Spectrometer for Femtosecond Electron Bunch Diagnostics by Single-Shot Spectroscopy of Coherent Radiation," *Nuclear Instruments and Methods A* (2011).
12. Q. Wu, and X.-C. Zhang "Ultrafast electro-optic field sensors " *Appl. Phys. Lett.* 68, 1604 (1996).
13. Y. Berozashvili, and et al., "Dispersion of the linear electro-optic coefficients and the non-linear susceptibility in GaP," *Journal of Physics D: Applied Physics* 22, 682 (1989).
14. S. Schulz, L.-G. Wissmann, J. Zemella, V. R. Arsov, M. Felber, P. Gessler, K. E. Hacker, F. Loehl, F. Ludwig, K.-H. Matthiesen, H. Schlarb, B. Schmidt, and A. Winter, "All-Optical Synchronization of Distributed Laser Systems at FLASH," in *Proceedings of the Particle Accelerator Conference 2009(Vancouver, Canada, 2009)*, pp. 4171-4173.
15. S. Schulz, M. K. Bock, M. Bousonville, M. Felber, P. Gessler, T. Lamb, F. Ludwig, S. Ruzin, H. Schlarb, and B. Schmidt, "Progress and Status of the Laser-based Synchronization System at FLASH," in *Proceedings of the European Workshop on Beam Diagnostics and Instrumentation for Particle Accelerators 2011(Hamburg, 2011)*.
16. M. K. Bock, M. Bousonville, M. Felber, P. Gessler, T. Lamb, S. Ruzin, H. Schlarb, B. Schmidt, and S. Schulz, "Benchmarking the Performance of the Present Bunch Arrival Time Monitors at FLASH," in *Proceedings of the European Workshop on Beam Diagnostics and Instrumentation for Particle Accelerators 2011(Hamburg, 2011)*.